Extended Corresponding States Equation of State for Natural Gas Systems[1]

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Abstract.

We present an extended corresponded states model for the prediction of thermodynamic properties of natural gas systems. This model is based on shape factors expressed as generalised functions of the reduced temperature and density, the acentric factor and the critical compression factor. A single set of coefficients was optimised for twelve major components of natural gases. The extension to multicomponent systems was carried out according to the one-fluid model with temperature- and density-dependent binary interaction parameters. Compression factors and speeds of sound of natural gases were predicted with average deviations within ±0.036 per cent.

Keywords: Equation of State, Corresponding states, Density, Compressibility factor, Speed of sound, Natural gas.

Introduction

Accurate prediction of the thermodynamic properties of natural gases is required for flow metering purposes under custody transfer and pipeline transmission conditions. Current standards call for the gas density r and speed of sound u to be predicted with uncertainty within ±0.1 per cent in the interval $273 \leq T/\mathrm{K} \leq 333$ K and $p/\mathrm{MPa} \leq 12$.

The thermodynamic modelling of natural gas systems represents a demanding compromise between accuracy and flexibility. The desired accuracy is currently achieved by pure-component equations of state (EoS) based on the Helmholtz free energy such as the recent reference equation for methane [1], but these models are not suitable for multicomponent applications. Extended corresponding states (ECS) models offer a viable option for they may be both highly accurate for pure fluids and applicable to mixtures. The accuracy of any ECS model depends upon proper enforcement of conformality between the mixture constituents and the reference fluid, application of a suitable mixture model and use of an accurate reference-fluid EoS. Several accurate ECS models have been reported in the last two decades including McCarty's model for liquefied natural gas [2], the model of Huber *et al* for refrigerants [3] and that of Clarke *et al* for the ternary system (N_2+Ar+O_2) [4].

The objective of this work was to develop an ECS model for natural gas under custody transfer conditions. This model should meet the required accuracy in both r and u in contrast to currently available models which cannot always achieve high accuracy in both thermal and caloric properties. For instance, the MGERG-88 model [5] is not suitable for caloric-property calculations and the AGA8-DC92 model [6] exhibits large deviations in u at high pressures and low temperatures when compared with experimental data [7,8].

Theory

Extended corresponding states relations

The corresponding states condition between two fluids is stated as follows:

$$Z(T, r) = Z_0(T/f, hr), \tag{1}$$

$$F^{res}(T, r) = F_0^{res}(T/f, hr). \tag{2}$$

Here, Z is the compression factor, $\Phi^{res} = A^{res}/nRT$ is the dimensionless residual Helmholtz free energy, and the subscript '0' refers to reference-fluid properties. The equivalent substance reducing ratios f and h map the properties of the fluid of interest onto those of the reference fluid. The simple Corresponding State Principle (CSP) asserts that the configurational properties of two fluids must be equal at the same reduced conditions; thus f and h are the simple ratios $f = T^c/T_0^c$ and $h = r_0^c/r^c$. This is valid only for small groups of very similar molecules such as the light noble gases. To extend the simple CSP to more complex systems, temperature- and density-dependent shape factors q and j are introduced such that,

$$f_{ii} = (T^c/T_0^c)q_{ii}(T_r, r_r), \tag{3}$$

$$h_{ii} = (r_0^c/r^c)j_{ii}(T_r, r_r). \tag{4}$$

Here, double subscripts are introduced to allow for extension to mixture nomenclature. It is possible to choose values of the shape factors such that equations (1) and (2) are obeyed exactly but, in practice, q and j are usually approximated by empirical correlations in terms of reduced temperature and density.

Application to mixtures

To apply the ECS formalism to mixtures, we adopt the van der Waals one-fluid model [9] whereby the configurational properties of the mixture are equated with those of a single hypothetical fluid. Thus, the configurational properties of the mixture can be calculated from those of the reference fluid as follows:

$$Z_x(T, r) = Z_0(T/f_x, h_x r), \tag{5}$$

$$F_x^{\text{res}}(T, r) = F_0^{\text{res}}(T/f_x, h_x r). \tag{6}$$

Here, the subscript 'x' refers to the mixture properties and f_x and h_x are the scaling parameters of the mixture which we obtain from the van der Waals mixing rules:

$$h_x = \sum_i \sum_j x_i x_j h_{ij}, \tag{7}$$

$$f_x h_x = \sum_i \sum_j x_i x_j f_{ij} h_{ij}. \tag{8}$$

The unlike terms which appear here are calculated from the conventional Lorentz-Berthelot combining rules with binary interaction parameters x_{ij} and h_{ij}:

$$f_{ij} = x_{ij}(f_{ii} \cdot f_{jj})^{1/2}, \tag{9}$$

$$h_{ij} = h_{ij}[\tfrac{1}{8}(h_{ii}^{1/3} + h_{jj}^{1/3})^3]. \tag{10}$$

As each of the mixture components is separately in corresponding states with the reference fluid, the arguments of equations (3) and (4) for f_{ii} and h_{ii} are $T_r = (Tq_{ii})/(f_x T_0^c)$ and $r_r = (h_x r)/(j_{ii} r_0^c)$.

Extended Corresponding States model

The first stage in the development of our ECS model was to obtain the behaviour of q and j for the main components of natural gas from experimental volumetric data on the pure substances. Since the method used to obtain q and j from experimental data is described in detail by Estela-Uribe [10], we give only a brief account here. The reference fluid was methane for which we used the equation of state of Setzmann and Wagner [1]. Experimental compression factors along isotherms were used to calculate F^{res}. To solve for q and j, equations (1) and (2) were expressed more conveniently as

$$Z(t,d) = Z_0(qt, jd), \tag{11}$$

$$F^{\mathrm{res}}(t,d) = F_0^{\mathrm{res}}(qt, jd). \tag{12}$$

Here, $t = T^{\mathrm{c}}/T$ is the inverse reduced temperature and $d = r/r^{\mathrm{c}}$ is the reduced density. Shape factors were obtained for ethane, propane, the two butanes, nitrogen, carbon dioxide and normal hydrogen. The data sources are listed in table 1. We next correlated q and j independently as functions of t and d and obtained generalised expressions for the parameters in terms of the acentric factor w and the critical compression factor Z^{c}. The shape factor correlations are:

$$q(t,d) = 1 + (w - w_0)\left(A_1(t) + A_2(t)\exp(-d^2) + Y_q(t,d)\right), \tag{13}$$

$$j(t,d) = (Z_0^{\mathrm{c}}/Z^{\mathrm{c}})\left\{1 + (w - w_0)\left(A_3(t) + A_4(t)\exp(-d^2) + Y_j(t,d)\right)\right\}. \tag{14}$$

Here, w_0 = 0.011406 is the acentric factor of methane calculated with the ancillary vapour pressure equation reported by Setzmann and Wagner [1]. The temperature-dependent coefficients are given by the general expression:

$$A_i(t) = a_{i,1} - a_{i,2}\ln t. \tag{15}$$

The functions Y_q and Y_j were included to improve the representation of the properties of pure substances in the critical region. They are given by

$$Y_q(t,d) = b_1 d \exp(-b_2 D^2), \tag{16}$$

$$Y_j(t,d) = c_1 d \exp(-c_2 D^2), \tag{17}$$

where D is a distance function defined by

$$\Delta(t,d) = (d-1)^2 + (t^{-1}-1)^2. \tag{18}$$

These correlations improved on those introduced by Leach *et al* [11]. Exact thermodynamic consistency is not observed by this method because each shape factor is correlated separately [12]. A method to enforce consistency was detailed by Estela-Uribe *et al* [13] and applied in the development of an alternative ECS model for natural gas mixtures [10]. However, the extent of the inconsistencies in the present model are quite small and we avoid them entirely by calculating all properties from $F^{\mathrm{res}}(t,d)$ and its partial derivatives.

The values of the coefficients in the model were obtained in a simultaneous fit to about 1500 data points comprising compression factors, vapour pressures and saturated vapour and liquid densities of ethane [14], propane [15], *i*-butane [16] and *n*-butane [17], and also speeds of sound of ethane [18]. Initially, literature values of w and Z^{c} were used but later w and Z^{c} were re-optimised as empirical constants on a substance-specific basis for the twelve most important components of interest. This improved the prediction of compression factors of components other than ethane and propane. Table 1 lists the coefficients. The shape-factor correlations are valid for the intervals $d \leq 2.5$ and $0.3 \leq t \leq 1.9$.

Temperature- and density-dependent binary interaction parameters were correlated as:

$$x_{ij} = (k_{ij,1} + k_{ij,2}T)(k_{ij,3} + k_{ij,4}r), \quad (19)$$

$$h_{ij} = (k_{ij,5} + k_{ij,6}T)(k_{ij,7} + k_{ij,8}r). \quad (20)$$

The coefficients k_{ij} were fitted to about 1300 binary-mixture Z and u data points. The volumetric data were taken from the GERG databank [19] for the systems given in table 2. The coefficients k_{ij} for the systems (CH_4+*i*-C_4H_{10}), (C_2H_6+C_3H_8), (C_2H_6+*n*-C_4H_{10}), (N_2+*i*-C_4H_{10}) and (CO_2+C_3H_8), for which there are no binary-mixture data reported in reference [19], were fitted to 980 natural-gas Z and u data points [19,7,8]. The acoustic data of Trusler *et al* were used for the systems (CH_4+C_2H_6) [20,8] and (CH_4+C_3H_8) [21], and data of Younglove *et al* [7] were used for the systems (CH_4+C_2H_6), (CH_4+C_3H_8), (CH_4+N_2), (CH_4+CO_2) and (N_2 + CO_2). We also included new speeds of sound measured in the mixtures (0.45783CH_4+0.55217N_2), (0.80037CH_4+0.19966N_2) and (0.80253CH_4+0.19747CO_2) at $170 \le T/\mathrm{K} \le 450$ K and $p \le 20$ MPa [10]. The k_{ij} values are given in table 2. The perfect-gas contribution to the heat capacities was taken from the generalised correlation of Jaeschke *et al* [22].

Results

We summarise the representation of pure-component compression factors in table 3. For the light hydrocarbons up to *n*-butane, the absolute average percentage deviations (AAD) are similar to, or better than, those obtained with MBWR equations of state [23]. The deviations in calculated Z of natural gases are given in table 3 for 4473 data points taken from reference [19] in the interval $270 \le T/\mathrm{K} \le 330$. The composition groups are those defined by GERG [5]. The root-mean-square (RMS) error or our deviations is

0.046 per cent whereas that reported by Jaeschke *et al* [5] for the MGERG-88 model is 0.055 per cent. Our results also compare favourably with the AAD of 0.035 per cent and the RMS error of 0.050 per cent reported by Savidge *et al* [6] for the AGA8-DC92 model. Figure 1 shows deviations in Z for a selected gas compared to those obtained with the two models mentioned above. Table 3 also gives the deviations in calculated u for 272 data points taken from four natural gases [7] and a synthetic gas [8] in the interval $275 \leq T/\mathrm{K} \leq 350$ and pressures up to 20 MPa for the gas of reference [8]. Our results compare quite favourably with those obtained with the AGA8-DC92 model for which we calculated an AAD of 0.056 per cent. Figure 2 contains deviations in calculated u for a selected gas. The overall AAD in calculated u for the three mixtures of reference [10] is 0.027 per cent with a maximum deviation of 0.14 per cent for the system ($0.80253CH_4+0.19747CO_2$) at 225 K and 3.35 MPa.

Conclusions.

The results we have obtained show that ECS models can achieve quite high accuracy in both density and sound speed when applied to either pure substances or multicomponent natural-gas mixtures. The range of applicability of the model may be extended if improved correlations of the binary interaction parameters can be found.

List of symbols.

a Coefficient in temperature-dependent term of shape-factor correlations.

A Helmholtz free energy or temperature-dependent term in shape-factor correlations.

b,c Coefficients of critical-enhancement functions in shape-factor correlations.

f,h *E*quivalent substance reducing ratios.

k Coefficient in binary interaction parameter correlation.

n Amount of substance.

p Pressure.

R Universal gas constant ($R = 8.31451\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}}$).

T Temperature.

V Volume.

x Mole fraction.

Z Compression factor.

Greek letters.

d Reduced density, r/r^{c}.

D Distance function.

h,x Binary interaction parameters.

q,j Shape factors.

r Amount-of-substance density. n/V

t Inverse reduced temperature, T^{c}/T.

F Dimensionless residual Helmholtz free energy, A^{res}/nRT.

Y Critical enhancement function.

w Acentric factor.

Superscripts.

c Critical property.

res Residual property.

Subscripts.

0 Reference-fluid property.

i,j Component indices.

x Mixture property.

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Table 1. Coefficients of shape-factor correlations.

Generalised coefficients		Substance-dependent coefficients		
		Component	w	Z^c
$a_{1,1}$	0.058990275	CH_4	0.011406	0.28629
$a_{1,2}$	0.930466257	C_2H_6	0.099349	0.27933
$a_{2,1}$	-0.088091565	C_3H_8	0.15124	0.27872
$a_{2,2}$	0.181659444	i-C_4H_{10}	0.18675	0.27787
$a_{3,1}$	0.004360921	n-C_4H_{10}	0.20086	0.27413
$a_{3,2}$	0.410925734	i-C_5H_{12}	0.227	0.27082
$a_{4,1}$	-0.644414001	n-C_5H_{12}	0.16082	0.26970
$a_{4,2}$	0.213433722	n-C_6H_{14}	0.299	0.26618
b_1	0.013017218	N_2	0.039988	0.28584
b_2	0.285879454	CO_2	0.21651	0.27391
c_1	-0.121476965	CO	0.062971	0.28651
c_2	0.123927229	H_2	-0.21515	0.31515

Table 2. Coefficients of binary-interaction-parameters correlations.

System	$k_{ij,1}$	$k_{ij,2}$/K^{-1}	$k_{ij,3}$	$k_{ij,4}$/dm^3·mol^{-1}	$k_{ij,5}$	$k_{ij,6}$/K^{-1}
CH_4+C_2H_6	1.040812	-0.000155	0.908914	-0.000004	1.077304	-0.003657
CH_4+C_3H_8	1.114644	-0.000620	1.009220	0.000338	1.333064	-0.011845
CH_4+*i*-C_4H_{10}	0.911592	0.000001	1.210727	0.000001	0.954675	0.000001
CH_4+*n*-C_4H_{10}	1.298363	-0.000520	1.043387	-0.000440	1.354056	-0.022930
CH_4+*n*-C_5H_{12}	1.254658	-0.000660	0.711106	0.000298	0.991373	-0.005850
CH_4+*n*-C_6H_{14}	1.176861	-0.001369	1.129391	0.000485	0.947194	0.009701
CH_4+N_2	1.012494	0.000266	1.046247	0.000018	1.061423	-0.003060
CH_4+CO_2	0.994786	-0.000427	1.168786	0.000419	1.293969	0.006469
CH_4+CO	1.302866	-0.001890	0.178940	0.001332	1.729690	-0.038778
CH_4+H_2	0.756318	0.001848	-1.106493	0.006492	0.923646	-0.012996
N_2+C_2H_6	1.184573	-0.000690	0.940269	-0.000410	1.212724	-0.017380
N_2+C_3H_8	0.927338	-0.000025	1.128330	-0.000130	1.012828	0.002439
N_2+*i*-C_4H_{10}	0.739760	0.000001	0.803401	0.000001	1.324881	0.000001
N_2+*n*-C_4H_{10}	1.051326	-0.001090	1.134815	-0.000490	1.448297	-0.023550
N_2+CO_2	1.330278	-0.001420	0.734213	0.000202	1.745215	-0.025700
N_2+CO	1.114119	-0.002760	0.752117	0.000287	2.407862	-0.071080
N_2+H_2	-0.222748	0.005248	1.849351	0.011969	0.959127	0.059379
CO_2+C_2H_6	0.757284	0.000417	1.378583	-0.000920	0.936519	0.001981

Table 2. –continued

System	$K_{ij,1}$	$k_{ij,2}$/K^{-1}	$k_{ij,3}$	$k_{ij,4}$/dm^3·mol^{-1}	$k_{ij,5}$	$k_{ij,6}$/K^{-1}
CO_2+C_3H_8	0.837776	-0.000886	1.493884	-0.000985	1.506068	-0.072175
C_2H_6+C_3H_8	1.021706	0.000103	0.715812	0.000388	0.992241	0.000882
C_2H_6+*n*-C_4H_{10}	0.788916	0.000001	1.368043	0.000001	1.095238	0.000001
C_2H_6+H_2	1.347633	-0.000520	0.775514	-0.000862	1.254104	-0.005140

System	$k_{ij,7}$	$k_{ij,8}$/dm^3·mol^{-1}	System	$k_{ij,7}$	$k_{ij,8}$/dm^3·mol^{-1}
CH_4+C_2H_6	0.997981	0.002435	N_2+C_3H_8	1.005513	-0.008248
CH_4+C_3H_8	0.651823	0.003173	N_2+*i*-C_4H_{10}	0.593652	0.000001
CH_4+*i*-C_4H_{10}	1.2131096	0.000001	N_2+*n*-C_4H_{10}	0.643310	0.015004
CH_4+*n*-C_4H_{10}	0.572448	0.007408	N_2+CO_2	0.564020	0.006511
CH_4+*n*-C_5H_{12}	0.970594	0.019257	N_2+CO	0.545059	0.012207
CH_4+*n*-C_6H_{14}	1.660691	-0.030591	N_2+H_2	0.022509	0.346004
CH_4+N_2	0.853525	0.003645	CO_2+C_2H_6	1.155950	0.008259
CH_4+CO_2	0.582983	0.000785	CO_2+C_3H_8	0.442717	0.075114
CH_4+CO	0.806935	0.013002	C_2H_6+C_3H_8	0.908270	0.013022
CH_4+H_2	0.612830	0.018448	C_2H_6+*n*-C_4H_{10}	1.026700	0.000001
N_2+C_2H_6	0.799955	0.015376	C_2H_6+H_2	0.744319	0.004332

Table 3. Statistical analysis of absolute average percentage deviations in compression factors and speeds of sound of pure components and natural gases.

AAD in compression factors of pure components					
Component	Overall	Super-critical	Near-critical	Sub-critical	Data source
C_2H_6	0.12	0.10	0.10	0.26	Ref. [14]
C_3H_8	0.06	0.04	0.08	0.04	Ref. [15]
i-C_4H_{10}	0.29	0.21	0.32	0.31	Ref. [16]
n-C_4H_{10}	0.33	0.29	0.36	0.37	Ref. [17]
n-C_5H_{12}	1.16	1.04	1.86		Ref. [24,25]
N_2	0.59	0.47	0.97	0.62	Ref[26,27,28]
CO_2	0.17	0.096	0.20	0.21	Ref[29,30,31]
CO	0.27	0.19	0.92	1.22	Ref. [32]
H_2	0.21	0.18	0.57		Ref. [33]

Deviations in compression factors of natural gases							
Gas	AAD	Bias	Max.Dev.	Gas	AAD	Bias	Max.Dev.
Group 1	0.040	-0.009	-0.16	Group 4	0.025	0.009	-0.13
Group 2	0.041	-0.022	-0.18	Group 5	0.031	0.010	0.17
Group 3	0.034	0.015	0.28	Group 6	0.027	0.010	-0.14
				Overall	0.033	0.005	0.28

Deviations in speeds of sound of natural gases							
Gas	AAD	Bias	Max.Dev.	Gas	AAD	Bias	Max.Dev.
Gulf Coast	0.016	0.003	0.061	Statvordgass	0.074	-0.008	0.37
Amarillo	0.014	-0.0003	0.072	Synthetic	0.043	0.032	0.15
Statoil Dry	0.049	-0.046	-0.13	Overall	0.036	-0.002	0.37

Figure captions.

Fig. 1. Relative deviations $\Delta Z/Z$ in compression factors Z of an Ekofisk natural gas (Group 3, N10) and those from the MGERG-88 and AGA8-DC92 models with respect to those calculated in this work. ▲ 273.15 K; — 273.15 K (MGERG-88); – – – 273.15 K (AGA8-DC92); ■ 293.15 K; ⋯ 293.15 K (MGERG-88); ----293.15 K (AGA8-DC92)

Fig. 2. Relative deviations $\Delta u/u$ in speeds of sound u of a Gulf Coast natural gas and those from the AGA8-DC92 model with respect to those calculated in this work. ▲ 275 K; — 275 K; ● 300 K; – – 300 K; ■ 350 K; ⋯ 350 K.

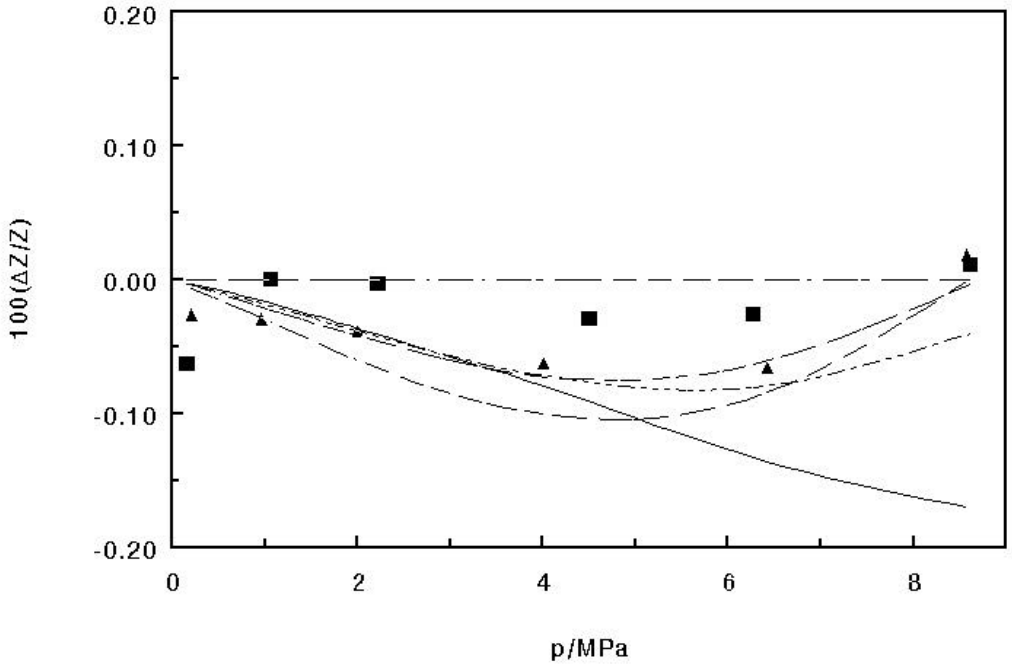

Figure 1.

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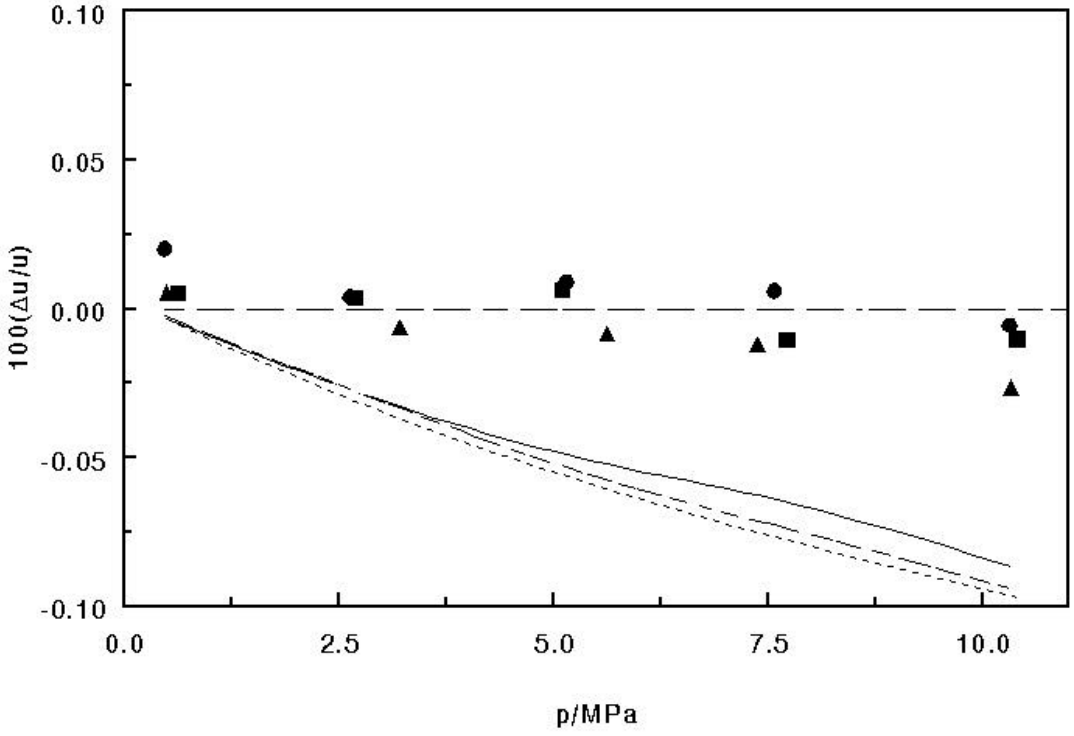

Figure 2.

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